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A Variety of Thorianite from Galle, Ceylon.

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In a previous paper* an account has been given of the composition and properties of a new mineral from Ceylon, chiefly composed of thorium dioxide (78 to 79 per cent.), to which the name thorianite was given. The mineral was shown to contain, besides thoria, a certain proportion of oxides of uranium varying in the three specimens analysed between 11 and 15 per cent., the uranium being present partly as dioxide and partly as trioxide, that is to say, in the same condition as in uraninite or pitchblende. It was shown that the evidence strongly supports the conclusion that thorianite and uraninite possess the same crystalline form and that they are isomorphous minerals. It is interesting to note that the new mineral naegite, from Japan, which is essentially a silicate of uranium containing thorium, appears to be isomorphous with thorite (thorium silicate). It was suggested that the intimate association of thoria with oxides of uranium in thorianite is to be regarded as a case of isomorphous mixture. Such a mixture, or perhaps "solid solution," might result from the crystallisation of thorianite from a fused mixture or magma containing the oxides of both elements.

We have now obtained important confirmation of this view through the analyses of some unusually large crystalline fragments of thorianite received from the Galle District of Ceylon, which show that a still wider variation may occur in the proportions of the two oxides present in the mineral. This variety is composed of black lumps, usually of indefinite crystalline form, some of the pieces being apparently cubic. They are often partially covered with a brownish yellow substance containing a considerable amount of silica and probably derived from the associated rock. Some of the pieces weighed over 50 grammes, and were evidently portions of much larger masses. The fractured surfaces often showed a slightly less lustrous and more pitchy appearance than ordinary thorianite. The appearance of the mineral is thus intermediate between the small separate and cubical crystals of thorianite and the large masses without definite crystalline form, which are usually characteristic of uraninite. A small amount of material from Hinidumpattu in Galle consisted of small cubic crystal almost indis-

* Dunstan and Blake, 'Proc. Roy. Soc.,' A, vol. 76, 1905.

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tinguishable in appearance from ordinary thorianite. In hardness, optical properties, density and general physical characters this variety of thorianite closely resembles the ordinary form of the mineral.

Analysis by the methods described in the previous paper* on thorianite shows, however, that it contains a much larger proportion (from 27 to 32 per cent.) of oxides of uranium and less oxide of thorium, whilst the minor constituents of the two forms of the mineral are seen to be similar in nature and quantity. It appeared possible that the masses might consist of uraninite crystallised on a nucleus of thorianite or *vice versa*, or that the two minerals might be separately crystallised within the masses. Analyses of different portions of one and the same piece have, however, given nearly identical results, and the oxides of the two elements appear to be uniformly distributed through the mineral, although there is some variation in the composition of specimens obtained by different collectors.

It may be noticed in the following table that the Analysis I of the small crystals from the Galle District shows rather more uranium and less thorium than that of the larger lumps (II to VI). If the uranium is calculated as UO_2 , the molecular ratio of ThO_2 to UO_2 in the small crystals is almost exactly 2 to 1, but there is no evidence that this is more than a coincidence. No. VII was a large crystal, 8 mm. cube, of ordinary thorianite from Balangoda.

The fact that thoria in thorianite is naturally associated with quantities of oxides of uranium varying from 11 to over 30 per cent., confirms the conclusions, indicated in the previous paper, that the oxides of thorium and uranium are present in thorianite in that intimate association known as "isomorphous mixture."

Different specimens of thorianite from the Galle district have furnished 58.84, 62.16, 62.30, 63.36 and 66.82 per cent. of thoria, whilst common thorianite from other localities has furnished from 76 to 79 per cent. of thoria.

The relations of thorium and uranium in minerals is a subject of some importance in connection with the present developments of the theory of the chemical elements. Attention has been directed recently to the subject by the Hon. R. J. Strutt.† It may be noticed in this connection that the percentages of thoria recorded by Mr. Strutt in his paper have been calculated from the observed radio-activity of the minerals, and not directly determined by chemical analysis. A comparison of the two series of percentages shows differences which in some cases are considerable. The calculated results are, however, only to be regarded as roughly approximate.

* *Loc. cit.*

† 'Proc. Roy. Soc.,' A, vol. 76, p. 88 1905.

The Galle variety of thorianite, as was to be expected, is radio-active, and it contains helium.

Mr. Strutt has kindly examined the radio-activity of this variety, and compared it with ordinary thorianite. The method of comparison was as follows:—"The quantity taken in each case was about one-tenth of a gramme. It was dissolved in strong nitric acid, and most of the excess of acid driven off by evaporation. The solution was diluted and exactly neutralised with ammonia, to prevent any injurious effect of the acid fumes on the apparatus. The solution was then made up to a standard volume.

The solutions of the two minerals were placed in exactly similar test-tube wash-bottles. These vessels were selected to have exactly the same diameter, and the inner tube in each case dipped to exactly the same depth in the solution. A continuous current of air could be drawn through either of them alternately into an electroscope. A two-way stop-cock made it easy to exchange one wash-bottle for the other. Constant suction was secured by the obvious device of letting in air through a deeper wash-bottle attached to a T-piece on the other side of the electroscope. The pressure driving the air through the electroscope was thus equal to the difference of depth between this bottle and the wash-bottle containing the active solution. The current was regulated once for all by a stop-cock, and was not sufficiently rapid to appreciably disturb the gold leaf of the electroscope.

In making the comparisons, air was first drawn through the solutions long enough to expel all accumulated radium emanation. Three measurements of the rate of leak due to the first solution were taken, then three with the other solution; then three more with the first, and so on. The mean of each of these sets was compared with that of its successor. The mean ratio so obtained was corrected for the slight difference in the quantities of material taken. The correction (or normal leakage of the electroscope) was too small to be worth applying. Two samples of each mineral were weighed out, and compared in the manner above described in every combination. Four values for the ratio of thorium activities in the two minerals were thus obtained. They were as follows:—

$$B/A = 1.19, 1.12, 1.10, 1.24. \quad \text{Mean, } 1.16.$$

The specimen of thorianite B used in this comparison contained 78.86 per cent. of thoria and 15.1 per cent. of uranium oxides. The specimen of the Galle variety A used was that numbered I in the table of analyses. The results of the radio-activity determinations are in fair accordance with the proportions of uranium and thorium determined by analysis."

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In the following table of analyses, I represents the composition of the small crystals from Hinidumpattu, Galle District; II to VI represent the compositions of the large lumps from the same locality; II, III, and IV are analyses of different parts of the same crystal, III being that of the outer layer; VII is an analysis of a large crystal of ordinary thorianite from Balangoda.

Table of Analyses.

	I.	II.	III.	IV.	V.	VI.	VII.
ThO ₂	58·84	62·16	66·82 {	—	62·32	63·36	78·98
(Ce,La,Di) ₂ O ₃	0·85	1·84		—	2·24	1·16	1·47
Y ₂ O ₃	—	—	—	—	—	—	—
UO ₂	32·74 {	10·32	28·24	28·68	27·02	27·99	13·40
UO ₃		18·88					
PbO	2·56	2·29	2·29	2·50	2·99	2·90	2·54
Fe ₂ O ₃	1·31	1·11	1·22	2·43	2·28	1·27	0·87
CaO	0·19	0·59	0·54	—	0·50	0·85	0·91
H ₂ O	1·26	1·05	1·00	—	2·16	1·32	1·28
Insoluble in nitric acid ...	0·45	0·77	0·56	0·54	0·87	0·77	0·47
He }	present	present	present	present	present	present	present
CO ₂ }							

Mineralogists have often assigned a new name to a mineral found in a new locality when it has differed essentially in composition from the previously known mineral. It does not, however, seem desirable, in the light of present knowledge, to regard this variety of thorianite from Galle as a new mineral species, especially when it is remembered that the oxides of thorium and uranium in thorianite are not chemically combined and that from the probable mode in which the mineral has been formed, it is to be expected that considerable variations would naturally occur in the proportions of the two oxides in the mineral found in distinct localities which have therefore crystallised under different conditions. In fact, it is probable that further examination of other specimens from distinct localities may reveal the existence of a series of substances intermediate between the hypothetical pure uraninite, consisting of uranium dioxide, and the pure thorianite, consisting of dioxide of thorium.